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Chemical contamination in southwest Puerto Rico: An assessment of organic contaminants in nearshore sediments

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Corals reefs are among the most productive and diverse ecosystems in the world, providing goods and services including commercial and subsistence fisheries, tourism and recreation, new medicines, and natural protection against storms for coastal communities and ports (Burke and Maidens, 2004; Waddell, 2005). Worldwide, however, coral reefs are experiencing significant degradation from increased nutrient load, sediments, and land-derived pollutants (Bryant et al., 1998; Spalding et al., 2001; Fabricius, 2005). Burke and Maidens (2004) noted that nearly two-thirds of coral reefs in the Caribbean are threatened by human activities. Furthermore, the US Coral Reef Task Force (USCRTF) identified pollution as a key threat to coral reef resources in Puerto Rico, the US Virgin Islands, and Southeast Florida (SEFCRI, 2004). Although pollution is frequently cited as a stressor on coral reefs, the occurrence and distribution of pollutants (chemical contaminants) in coral reefs have not been well characterized. Quantifying the types and levels of chemical contaminants in coral reef ecosystems is an essential step toward understanding their impacts, and ultimately mitigating their effects. In most coral reef systems, managers do not have adequate information on the concentration and spatial distribution of contaminants, which in some areas may be critical for the successful and sustained management of reef resources. This paper contains a spatial characterization of organic chemical contaminants occurring in sediments adjacent to coral reefs and hardbottom substrates in the study area in southwest Puerto Rico, and is part of a larger study to examine linkages between chemical contamination and coral condition (Pait et al., 2007).

The study area in southwest Puerto Rico was approximately 20 km long, and included the nearshore waters adjacent to the town of La Parguera and then east to Guanica Bay (Fig. 1). In this part of Puerto Rico, the shelf area extends approximately 10 km from shore, then drops off at about 20 m depth. The shelf has both emergent and submerged reefs and contains a variety of hard and soft corals, along with extensive seagrass beds, coastal fringing mangroves and a series of mangrove islands (Kendall et al., 2001; Christensen et al., 2003). The predominant longshore flow in the study area is from east to west (CFMC, 1998). The town of La Parguera has a population of approximately 26,000 and is a popular weekend and vacation destination. Guanica Bay in the eastern portion of the study area contains the towns of Ensenada and Guanica, and over the years several industrial operations including sugar processing, fertilizer mixing and textile manufacture have operated there. In addition, the Lajas Valley (Fig. 1) which has a significant amount of agricultural production, drains to Guanica Bay through a series of manmade canals.

For this study, a stratified random sampling design was used to select the sampling points. Determining the location of sediments in the study area was made possible by

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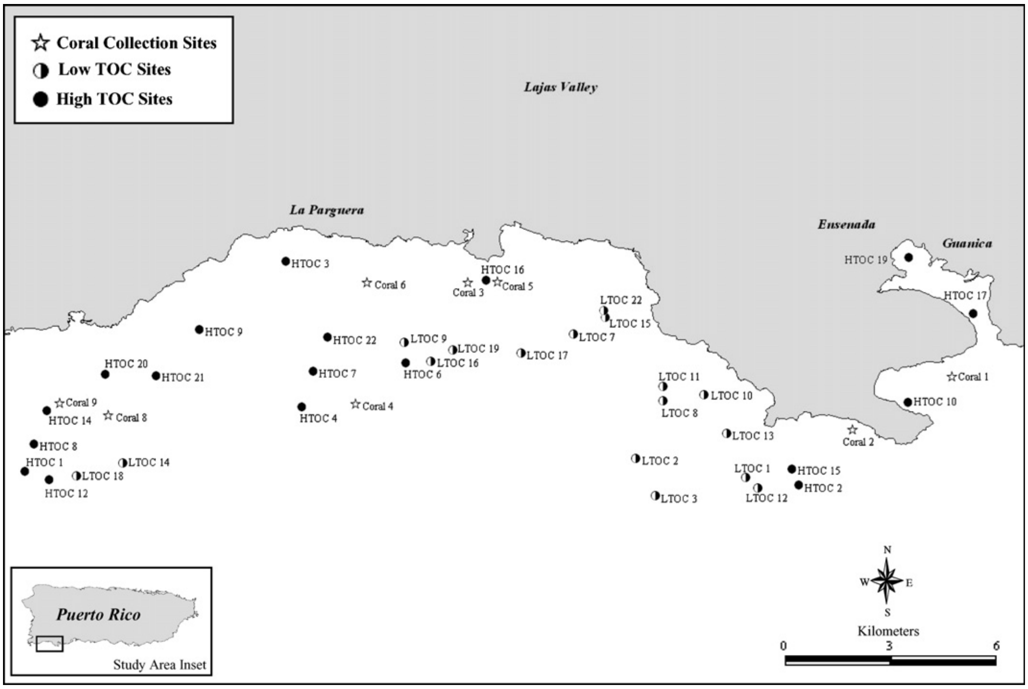

Fig. 1. Sediment sampling sites in southwest Puerto Rico. Low-TOC (LTOC), low modeled total organic carbon sites; High-TOC (HTOC), high modeled total organic carbon sites; Coral, sites with sediments adjacent to corals.

the extensive benthic habitat mapping completed by NOAA's Center for Coastal Monitoring and Assessment (CCMA) (Kendall et al., 2001). Using ArcGIS®, the hard-bottom (coral) data layer was removed leaving the soft bottom sediment layer which was then stratified into mangrove, lagoon, backreef and bankshelf zones. A preliminary survey of the sediments was used to characterize and then model sediments for higher (HTOC) and lower (LTOC) total organic carbon. The sampling points for this study (Fig. 1) were then randomly selected using ArcGIS®.

Sediment samples were collected in August 2005 using protocols developed by the National Status and Trends (NS&T) Program (Lauenstein and Cantillo, 1993) which resides in CCMA. A modified Van Veen grab was deployed to collect sediment samples, and the top 3 cm of sediment were removed from the grab using a Teflon coated scoop and placed into a 250 ml certified clean glass container. All samples for organic chemical analyses were placed on ice and then frozen at the end of each day. A sample of sediment was also taken and refrigerated for grain size analysis. PAHs were analyzed using gas chromatography/mass spectrometry in the selected ion monitoring mode. PCBs and organochlorine pesticides were analyzed using gas chromatography/electron capture detection. Butyltins were analyzed using gas chromatography/flame photometry. Detailed analytical protocols for the organic compounds can be found in Kimbrough et al. (2006). The PAHs, PCBs, organochlorine pesticides and butyltins analyzed as part of this project are included in Table 1. A subsequent publication will present the results of the major and trace element analyses.

All contaminant data were analyzed using JMP® statistical software. A Shapiro-Wilk test was first run on individual parameters to see if the data were normally distributed. When the data were not normally distributed and transformations were not effective, Spearman's nonparametric multivariate correlation was used. Differences in contaminant classes among sample strata (habitat type) also were investigated. ANOVAs were run when tests (O'Brien's test) revealed adequate homogeneity of variance on the transformed data. Pairwise comparison tests (Tukey-Kramer HSD) were then run to assess differences among means.

The average depth of the sites sampled was 10.3 ± 6.0 m, average salinity 34.2 ± 0.7 ppt, and the average surface water temperature was 30.2 ± 0.4 °C. The characteristics of the sediments by habitat type are shown in Table 2. The lagoon areas had the highest silt/clay (52%) composition, and the percent total organic carbon (TOC) for all habitat types was roughly 1%.

Total PAHs in this paper refers to the sum of the 24 PAHs highlighted in Table 1. Fig. 2 shows the concentration of total PAHs measured in the sediments. Total PAHs at the majority of sites sampled were below 300 ng/g. Somewhat elevated concentrations of total PAHs were found in the sediments adjacent to the town of La Parguera, with even higher concentrations detected at the two sites sampled in Guanica Bay. Because of the long-term, national-level contaminant monitoring carried out by the NS&T Program, data from southwest Puerto Rico can be compared with NS&T results from the rest of the Nation's coastal waters over the last 20 years. The two sites in Guanica Bay, HTOC 17 (583 ng/g) and HTOC 19 (911 ng/g),

Table 1
Selected organic chemical contaminants quantified in the sediments from southwest Puerto Rico

PAHs	PAHs	PCBs	Organochlorine pesticides
Naphthalene*	Chrysene*	PCB8/5*	Aldrin
C1-Naphthalenes	C1-Chrysenes	PCB18*	Dieldrin
C2-Naphthalenes	C2-Chrysenes	PCB28*	Endrin
C3-Naphthalenes	C3-Chrysenes	PCB29	Heptachlor
C4-Naphthalenes	C4-Chrysenes	PCB31	Heptachlor-Epoxide
Benzothiophene	Benzo(*b*)fluoranthene*	PCB44*	Oxychlordane
C1-Benzothiophenes	Benzo(*k*)fluoranthene*	PCB45	Alpha-Chlordane
C2-Benzothiophenes	Benzo(*e*)pyrene*	PCB49	Gamma-Chlordane
C3-Benzothiophenes	Benzo(*a*)pyrene*	PCB52*	Trans-Nonachlor
Biphenyl*	Perylene*	PCB56/60	Cis-Nonachlor
Acenaphthylene*	Indeno(l,2,3-*c*,*d*)pyrene*	PCB66*	Alpha-HCH
Acenaphthene*	Dibenzo(*a*,*h*)anthracene*	PCB70	Beta-HCH
Dibenzofuran	C l-Dibenzo(*a*,*h*)anthracenes	PCB74/61	Delta-HCH
Fluorene*	C2-Dibenzo(*a*,*h*)anthracenes	PCB87/115	Gamma-HCH
Cl-Fluorenes	C3-Dibenzo(*a*,*h*)anthracenes	PCB95	2,4′-DDD
C2-Fluorenes	Benzo(*g*,*h*,*i*)perylene*	PCB99	4,4′-DDD
C3-Fluorenes		PCB101/90*	2,4′-DDE
Carbazole	*Individual PAH isomers*	PCB105*	4,4′-DDE
Anthracene*	2-Methylnaphthalene*	PCB110/77	2,4′-DDT
Phenanthrene*	1-Methylnaphthalene*	PCB118*	4,4′-DDT
C1-Phenanthrene/anthracenes	2,6-Dimethylnaphthalene*	PCB128*	1,2,3,4-Tetrachlorobenzene
C2-Phenanthrene/anthracenes	1,6,7-Trimethylnaphthalene*	PCB138/160*	1,2,4,5-Tetrachlorobenzene
C3-Phenanthrene/anthracenes	1-Methylphenanthrene*	PCB146	Hexachlorobenzene
C4-Phenanthrene/anthracenes		PCB149/123	Pentachloroanisole
Dibenzothiophene		PCB151	Pentachlorobenzene
C1-Dibenzothiophenes		PCB153/132*	Endosulfan II
C2-Dibenzothiophenes		PCB156/171/202	Endosulfan I
C3-Dibenzothiophenes		PCB158	Endosulfan sulfate
Fluoranthene*		PCB170/190*	Mirex
Pyrene*		PCB174	Chlorpyrifos
C1-Fluoranthenes/pyrenes		PCB180*	
C2-Fluoranthenes/pyrenes		PCB183	*Butyltins*
C3-Fluoranthenes/pyrenes		PCB187*	Monobutyltin
Naphthobenzothiophene		PCB194	Dibutyltin
C1-Naphthobenzothiophenes		PCB195/208*	Tributyltin
C2-Naphthobenzothiophenes		PCB201/157/173	Tetrabutyltin
C3-Naphthobenzothiophenes		PCB206*	
Benz(*a*)anthracene*		PCB209*	

* Compounds used in the calculation of total PAHs or total PCBs.

Table 2
Mean values of sediment characteristics by habitat for sampling sites in southwest Puerto Rico

Zone	Number of samples	%Silt/clay	%Total organic carbon	Depth (m)
Coral	8	16.0 ± 4.0	1.4 ± 0.5	9.7 ± 1.7
Lagoon	10	52.0 ± 11.2	1.0 ± 0.2	6.7 ± 1.0
Backreef	6	5.2 ± 0.5	1.1 ± 0.3	1.6 ± 0.2
Bankshelf	19	34.9 ± 7.7	1.2 ± 0.3	15.2 ± 0.8

Values represent means ± SE.

were the only sites in the study area above the national NS&T median of 415 ng/g for total PAHs. None of the sites sampled in southwest Puerto Rico approached the national NS&T 85th percentile of 2688 ng/g.

Habitats in the study area vary from nearshore mangrove and lagoon sites to shallow areas behind emergent reefs (backreef), to the deeper bankshelf reefs. To assess whether total PAHs ($\log_{10}$ transformed) in the sediments varied by habitat, an ANOVA was run followed by pairwise comparisons (Tukey-Kramer HSD). The results indicated a significant difference ($P > 0.0077$, $r^2 = 0.2605$) in total PAH concentration among habitats. The pairwise comparison indicated that the concentrations of total PAHs in the backreefs (0.58 ± 0.29 ng/g) were significantly lower than in the lagoon (1.89 ± 0.23 ng/g) areas.

The adsorption of organic contaminants onto sediments is strongly influenced by grain size. A regression run between grain size and the concentration of total PAHs in the sediment samples indicated a highly significant ($P < 0.0001$, $r^2 = 0.7694$) relationship between the silt/clay fraction of the sediment and the concentration of total PAHs (Fig. 3). Interestingly, there was no significant relationship between TOC and the concentration of total PAHs ($P > 0.6531$, $r^2 = 0.005$), as has been found by others (Shine and Wallace, 2000; Hassett et al., 1980) in freshwater and estuarine systems. The lack of a relationship may have been related to the low TOC values encountered in the study area.

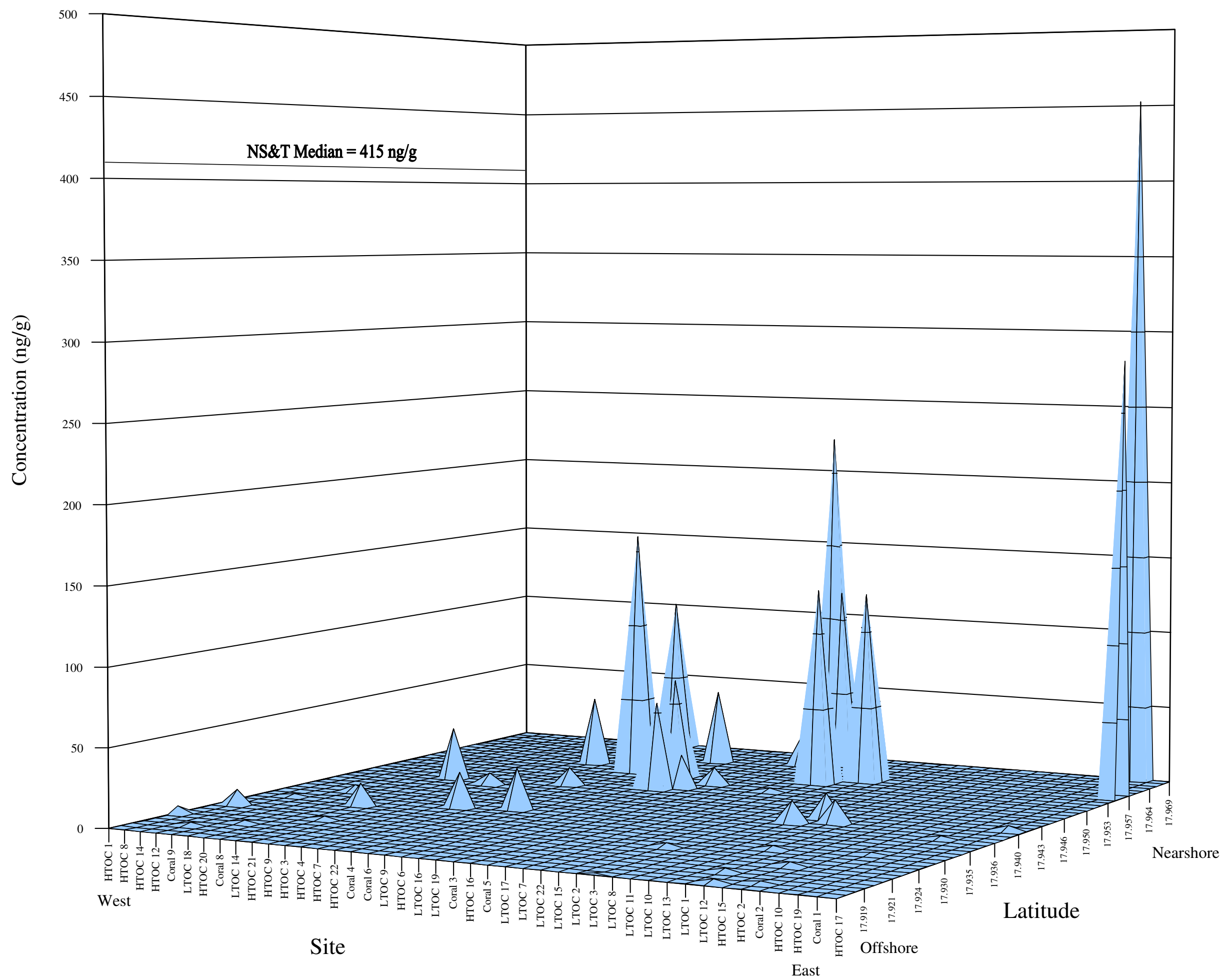

Fig. 2. Total PAHs detected in the sediments in southwest Puerto Rico. The sites are organized in a west to east direction to be comparable with Fig. 1. Higher latitudes are generally associated with nearshore locations in the study area.

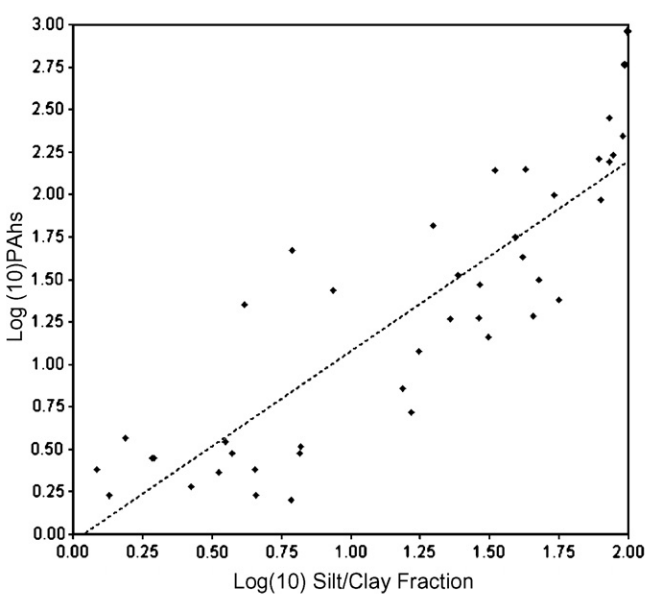

Fig. 3. Plot of log10 normalized %silt/clay (fines) and log10 normalized total PAHs detected in the sediments in southwest Puerto Rico. Dotted line represents the linear regression (log10 total PAHs = −0.041562 + 1.119682 log10 silt/clay fraction).

The distribution of high ($\geqslant$4 rings) molecular weight (HMW) versus low molecular weight (LMW) PAHs in sediment samples has been used as an indicator of pyrogenic versus petrogenic sources (Neff et al., 2005). An analysis of the sediment data from southwest Puerto Rico revealed no significant difference in the mix of LMW and HMW PAHs (Prob > ChiSq = 0.4472) in the sediment samples, which would indicate a mixture of both petrogenic and pyrogenic sources of the PAHs. An analysis of nonalkylated versus alkylated (e.g., C1-napthalenes) compounds, however, indicated the concentration of nonalkylated PAHs was significantly higher (Prob > ChiSq = 0.0422). This could be indicative of a higher proportion of pyrogenic PAHs, but could also be the result of a limited number of alkylated PAHs analyzed in the samples.

Researchers including Kimbrough and Dickhut (2006), Alsberg et al. (1985), Rogge et al. (1993), Singh et al. (1993), and Khalili et al. (1995) have investigated the signatures from various sources including automobile exhaust, road dust, coal dust and creosote to fingerprint various types of combustion PAHs. Kimbrough and Dickhut (2006) presented patterns of five high molecular weight

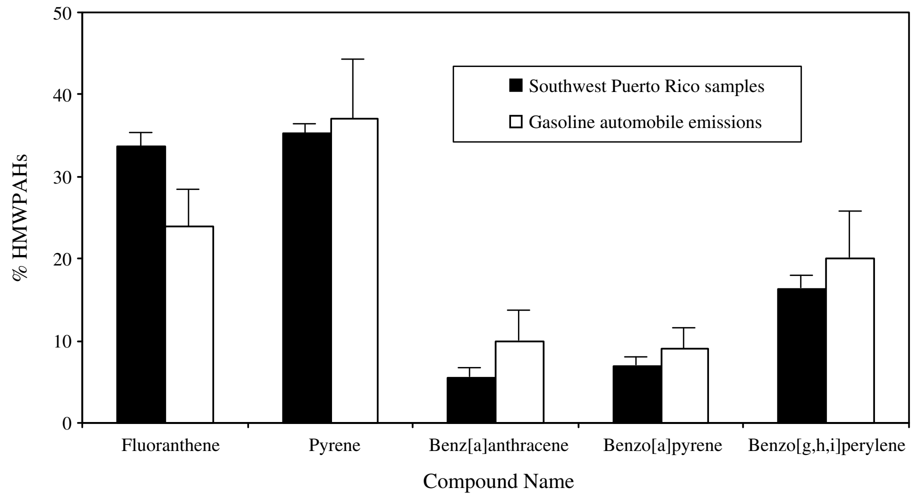

Fig. 4. Comparison of high molecular weight (HMW) PAHs in the sediment samples from southwest Puerto Rico with emissions from gas automobile engines. Data on automobile engine emissions from: Kimbrough and Dickhut (2006); Alsberg et al. (1985), Rogge et al. (1993), Singh et al. (1993), and Khalili et al. (1995). %HMW PAHs = 100 * [HMW PAH concentration/∑fluoranthene, pyrene, benzo[*a*]anthracene, benzo[*a*]pyrene, benzo[*ghi*]perylene]. Error bars represent standard error.

PAHs including fluoranthene, pyrene, benzo[*a*]anthracene, benzo[*a*]pyrene, and benzo[*ghi*]perylene from various sources. Fig. 4 contains a comparison of selected high molecular weight (HMW) PAHs in the sediment samples from southwest Puerto Rico expressed as a percent, to emissions from automobiles. It can be seen from Fig. 4 that the pattern of these five HMW PAHs from the sediments appears similar to the automobile engine emission signature, indicating that automobile emissions may be an important source of PAHs in the sediments in the study area. Miller et al. (2003) assessed the PAH signature from various types of outboard engines for boats. They found that outboard engine exhaust was enriched in acenapthylene, fluorene and phenanthrene for all outboard engine types tested. The sediments collected in southwest Puerto Rico, however, had higher relative contributions of fluoranthene and pyrene. Although recreational boating is a popular activity in the study area, the pattern of PAHs in the sediments appears more similar to emissions from automobile than outboard engines.

Total PCBs detected in the sediments in southwest Puerto Rico are shown in Fig. 5. Total PCBs as presented is the sum of the 18 congeners highlighted in Table 1. As with the PAHs, there was a pattern of elevated total PCB levels adjacent to La Parguera and especially in Guanica Bay.

NOAA's NS&T Program developed effects-based, numerical guidelines to estimate the toxicological relevance of certain sediment contaminants (Long et al., 1998). These guidelines, the Effects Range-Low (ERL) and the Effects Range-Median (ERM) define contaminant concentration ranges in the sediments that are rarely (<ERL), occasionally (ERL–ERM) or frequently (>ERM) associated with toxic effects (NOAA, 1998). The ERL and ERM values for total PCBs are also included in Fig. 5.

Several sites around La Parguera, including HTOC 16, HTOC 22, and LTOC 22 all had total PCB levels somewhat above the ERL of 22.7 ng/g. In Guanica Bay however, the concentration of total PCBs at the two sites sampled was not only above the ERL, but substantially above the ERM value of 180 ng/g. Total PCBs at HTOC 19 was 469 ng/g, and at HTOC 17, a concentration of 1243 ng/g was found. The presence of substantially elevated levels of PCBs in Guanica Bay could be the result of a spill, industrial discharges, dumping of PCB contaminated equipment (e.g., transformers), discharges from ships, or runoff (e.g., from rainfall or cleaning activities) from industrial sites, to mention a few possibilities. As noted, there have been a number of industrial operations around Guanica Bay over the years. Because only two sediment samples were taken within the Bay as part of the larger overall sampling design, it is not possible to statistically characterize PCB levels in Guanica Bay from this study. Additional sampling would be required in order to characterize PCBs and other contaminants within Guanica Bay.

The relationship between PCBs and grain size also was examined. A nonparametric analysis of the data revealed a significant positive correlation between the silt/clay fraction of the sediment, and total PCBs ($P < 0.0001$, Spearman Rho = 0.8584).

As with PAHs and PCBs, there were elevated levels of total DDT in the sediments near the town of La Parguera and at the two sites in Guanica Bay. With the exception of Guanica Bay, concentrations of total DDT were below the ERL value of 1.58 ng/g. At the two sites in Guanica Bay, total DDT exceeded the NS&T 85th percentile of 18 ng/g, and at HTOC 17, a total DDT concentration of 46.9 ng/g slightly exceeded the ERM value (46.1 ng/g), indicating a greater likelihood of biological effects on biota

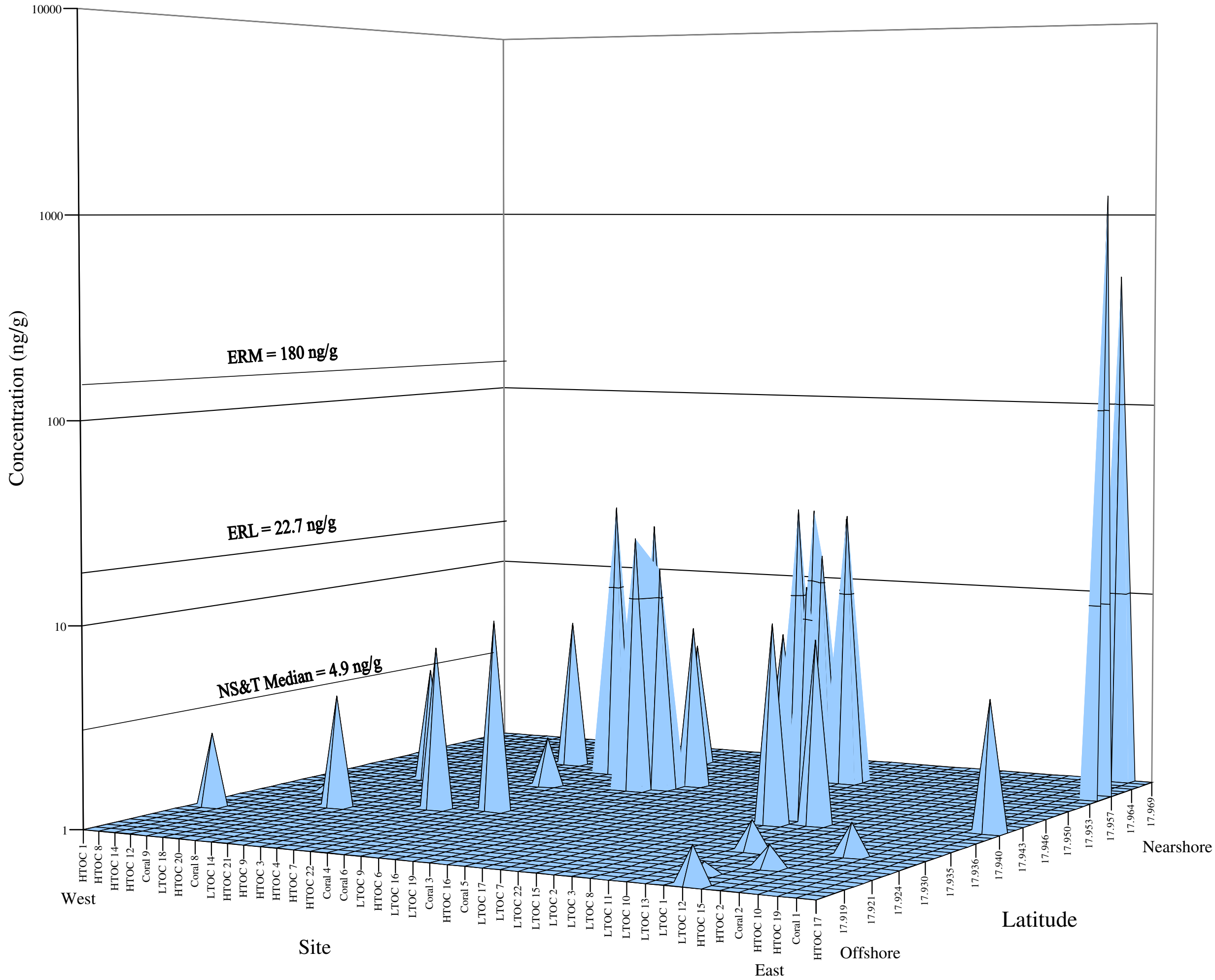

Fig. 5. Total PCBs detected by site in the sediments in southwest Puerto Rico. The ERL or Effects Range-Low is the concentration of a contaminant or contaminant class below which toxicity (10th percentile) is rarely observed. The ERM or the Effects Range-Median is the concentration above which toxicity in test organisms is more frequently (50th percentile) observed. Higher latitudes are generally associated with nearshore locations in the study area.

Table 3
Summary of results for other selected contaminant classes

Contaminant class	Number of samples	Mean (ng/g)	Number above NS&T 85th percentile
Total DDT	43	2.10 ± 1.26	2
Total chlordane	43	0.15 ± 0.06	1
Total dieldrin	43	0.03 ± 0.01	0
Tributyltin	43	0.01 ± 0.01	0

Mean ± SE.

at the site. As with total PCBs, however, elevated levels of total DDT did not appear to be present just outside of Guanica Bay. A regression of total DDT (log10 transformed) and the silt/clay fraction followed by a nonparametric analysis indicated a significant positive correlation ($P < 0.0001$, Spearman Rho $= 0.8301$). The elevated levels of DDT and its metabolites in Guanica Bay could be related to past agricultural activity. The Lajas Valley has had extensive agricultural activity over the years, including sugarcane and pineapple production. DDT was likely applied in the past to both of these crops to control insects on the foliage and in the soil.

A number of other organochlorine pesticides included in this study were also detected in the sediments including total dieldrin (sum of dieldrin and aldrin), endrin, and total chlordane. None of these compounds or compound classes currently have ERL or ERM values. Most concentrations were below the NS&T national median (Table 3).

Mono-, di-, tri-, and tetrabutyltin in the sediments were also analyzed as part of this project. TBT, used in antifoulant paints for boat hulls, was banned for use in the US on most smaller boats in 1989. There were few detections of tributyltin (TBT) in the sediments sampled in southwest Puerto Rico. Detectable TBT was only found at three sites, two of which were in Guanica Bay. The highest concentration of TBT found was 0.29 ng/g at HTOC 3. At all three sites, TBT concentrations were below the NS&T national median of 0.41 ng/g. Monobutyltin was the dominant butyltin found, and is likely the result of TBT debutylization. Several sites (HTOC 12, HTOC 21, LTOC 16, and LTOC 22) had sediment concentrations above the NS&T national median for monobutyltin (0.26 ng/g). The highest monobutyltin concentration found in the sediments in the study area was 1.54 ng/g at HTOC 3, near the town of La Parguera (Fig. 1).

The degradation of coral reef ecosystems worldwide has led to intensive efforts to understand and mitigate the stressors responsible for the declines of these valued and fragile ecosystems. The role of pollution in the degradation of coral reefs is often cited as a major factor, but the degree to which pollution and more specifically chemical contaminants are present and impact coral reefs is in most cases unknown. Because of this, coral reef managers may be missing an important piece of information needed to effectively manage reef areas. Quantifying the types and concentrations of chemical contaminants present in coral reef habitats is an important step in understanding possible impacts at the community and individual levels in corals and other reef inhabitants.

Overall, the levels of chemical contaminants in the sediments in the study area were fairly low. At most locations, sediment contaminant concentrations were less than the national median values in NOAA's NS&T Program, which monitors chemical contamination in all coastal waters of the US. A pattern seen in the study area for a number of contaminant classes included somewhat elevated concentrations adjacent to the town of La Parguera with even higher concentrations at the two sites sampled in Guanica Bay. For total PCBs, the ERM or Effects Range-Median was exceeded at both sites in Guanica Bay. These results provide a baseline characterization of the spatial distribution of contaminants in sediments adjacent to coral reefs upon which an ecosystem approach could be developed to understand and mitigate the effects of contaminants in coral reef ecosystems in southwest Puerto Rico.

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BITS: A SMART indicator for soft-bottom, non-tidal lagoons

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The implementation of the European Water Framework Directive (WFD, 2000/60/EC) has provoked a huge debate about the use of benthic indices to determine the quality of European coastal and estuarine waters (Magni et al., 2004; Borja, 2006; Dauvin, 2007). The biotic indices AMBI (Borja et al., 2000), BENTIX (Simboura and Zenetos, 2002), and BQI (Rosenberg et al., 2004) were created to classify the different coastal and estuarine water masses in accordance with the WFD. These indices are based on taxonomic identification at the species level, except for Oligochaetes which are identified at the class level. As Dauvin (2005) recently pointed out, "the WFD seeks to establish biological indicators for evaluating the quality of littoral milieus, but how can such indicators be established and proposed by those with real no expertise, and without any certitude that the species identified along the latitudinal lines of the European Union are really the same?" To overcome this problem, Dauvin and Ruellet (2007) proposed the BOPA (the Benthic Opportunistic Polychaetes Amphipods index) whose main innovation was the application of the "taxonomic sufficiency" principles (Ferraro and Cole, 1990) to an index, thus minimizing the number of identification errors (Dauvin and Ruellet, 2007). Recently, Munari and Mistri (2007a) explored the usefulness of two of such indices (AMBI and BOPA) to their use in Mediterranean transitional water ecosystems, in determining the benthic status according to the WFD (BQI was not used since it does not seem to be applicable to very shallow lagoons, where depth rarely exceed 1.5 m; BENTIX was specifically developed for the Mediterranean Sea, but Simboura and Zenetos (2002) advised against its use in transitional waters, where the natural conditions favour the presence of tolerant species in very high densities). Different indices rendered very different results, and just in a few cases a good agreement on the ecological quality (EcoQ) of a station was obtained: different but non-consistent responses of the different indices might lead to doubt in managers' minds regarding the value of the methods, and can produce confusion regarding whether remediation measures are needed.

Coastal lagoons are characterised by highly variable physico-chemical and hydro-morphologic conditions, typically resulting in a mosaic of different habitats. Due to the high variability of environmental parameters in the lagoons (e.g. salinity, dissolved oxygen, temperature), the species living in such environments adapt to the variability (Cognetti, 1992) and become tolerant of changes (Cognetti and Maltagliati, 2000). Lagoons show a relative paucity of species with respect, e.g. to the marine environment (Fredj et al., 1992). Coastal lagoons are naturally organic rich systems, being structurally eutrophic habitat islands in the coastal landscape. Given this specificity, it would seem prudent to develop specific methods for coastal lagoons before implementing the WFD in transitional waters. Within this framework, we developed the new index BITS (a Benthic Index based on Taxonomic Sufficiency) specifically for coastal lagoons in the Mediterranean Sea.

We used quantitative data regarding the composition and abundance of macrofauna collected during 32 surveys between 1999 and 2005 from six coastal lagoons located

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